# Study of the Origin of Misorientation in GaN Grown by Pendeo-Epitaxy

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## ABSTRACT

Growth of pendeo-epitaxial (PE) layers introduces misorientation between the seed layers and the overgrown wing layers. The origin of this misorientation has been studied by Transmission Electron Microscopy (TEM) using a set of samples in which subsequent procedures utilized in PE were applied, i.e. growth of GaN template, stripe etching, annealing at the growth temperature of the PE layers and final PE growth. It was shown that etching of seed-stripes did not change the type of defects or their distribution. However, heating to the PE growth temperature drastically modified the surface and V-shaped pits were formed. The surface became smooth again after the PE growth took place. Overgrowth of the V-shaped pits resulted in formation of edge threading dislocations over a seed-stripe region with a dislocation density of $8.0 \times 10^8$ cm$^{-2}$. Formation of new edge dislocations over the seed can have an influence on the misorientation between the PE grown regions.

## INTRODUCTION

GaN layers are usually grown on sapphire or SiC substrates with either an AlN or a GaN buffer layer. The large misfit in lattice parameters and in thermal expansion coefficients results in a high density of misfit and threading dislocations in the GaN layers ($10^9$-$10^{10}$ cm$^{-2}$). Although light emitting diodes with high efficiency have been produced despite the high dislocation density in this material, other applications such as lasers require dislocation reduction. Methods to reduce dislocation density include lateral epitaxial overgrowth (LEO) [1-3] or pendeo-epitaxy (PE) [4-6]. Both these methods result in a much lower dislocation density in the overgrown areas, but at coalescence fronts (CF) the dislocation density often exceeds or is comparable to the dislocation density observed using conventional growth on SiC or $Al_2O_3$ substrates. Misorientation (such as tilt/twist) between the seed and overgrown wings has been reported [3, 6-8]. Typical values of misorientation are 1-2° for LEO or PE with mask applied on the seed-stripes and 0.05-0.3° for mask-free PE. In order to further reduce dislocations in material grown by PE and understand what leads to the wing misorientation, structural studies were performed for each step of the PE process using TEM.

## EXPERIMENTAL

1 µm GaN layers were grown by metalorganic vapor phase epitaxy on 6H-SiC (0001) substrates with a 0.1 µm thick AlN buffer layer. GaN stripes along [1$\underline{1}$00] were fabricated by etching through the epitaxial layers into the substrate with an inductively coupled plasma. The stripe width and the stripe period were 3-5 µm and 5-12 µm, respectively. Subsequent mask-free

pendeoepitaxial growth was performed (Fig.1) at temperatures between 1080 and 1100 °C using a V/III ratio of 6200 and 1300, respectively. These growth conditions resulted in either coalesced or uncoalesced films. Further details on the growth process can be found in [9].

The set of samples from different stages of PE growth included: GaN template, followed by samples which underwent the etching procedure to form the stripes, and then subsequently samples where annealing for 10 min at 1000 °C and 20 Torr under a mixture of ammonia and hydrogen was performed. The annealing atmosphere corresponded to the standard gas flow conditions used for PE growth, whereas the annealing temperature was slightly lower than the PE growth temperature to limit the degradation of the GaN surface due to decomposition. The annealing time is significantly longer than the time used to stabilize the temperature prior to the PE growth, and it is assumed to be sufficiently long to simulate the thermal load on the stripes during PE growth. Finally samples with PE growth were also studied.

Samples were studied by transmission electron microscopy (TEM) using a JEOL 3010 and Topcon 002B microscopes operated at 300kV and 200kV respectively. Cross-sectional samples for TEM were prepared in a direction perpendicular to the seed-stripe. A standard bright or dark field method for determination of dislocation Burgers vectors was utilized.

## RESULTS AND DISCUSSION

The starting GaN sample used for the PE process had a smooth surface (Fig.2a). Bright field images in two-beam conditions obtained for two perpendicular sets of diffraction vectors ($g$) revealed the presence of pure screw (s) and pure edge (e) dislocations. Occasionally mixed-type dislocations (m) were observed. In Fig.2a, taken with $g=\underline{11}20$, where edge and mixed dislocations should be in a contrast, much higher dislocation density was observed in the area close to the GaN/AlN interface compared to the area near the GaN surface, as observed earlier [10]. The dislocation density measured within the GaN thicknesses $0<t<0.5\mu m$ was $19\times10^{8}cm^{-2}$, whereas for thicknesses $0.5<t<1.0\mu m$ it was $5.4\times10^{8}cm^{-2}$.

The next step in the PE process was etching of seed-stripes. As a result seed-stripes with width of 4.9μm and separation of 5.5μm were obtained. The etching did not change the GaN layer thickness, or the surface morphology and dislocation density (Fig.2b).

The sample with etched stripes was used for further growth of GaN. In order to learn if this process step introduces any changes to the GaN seeds via the thermal load during PE growth, the sample with etched stripes was annealed for 10min at 1000°C. TEM studies of a cross-section sample show that the annealing process drastically changed the surface morphology of the GaN seeds. The surface became rough and V-shaped pits were formed. The peak-to-valley roughness of the GaN surface was about 150nm (Fig.2c). The angle between walls of the V-shaped pits was measured to be about 60°. This value is very close to the angle of
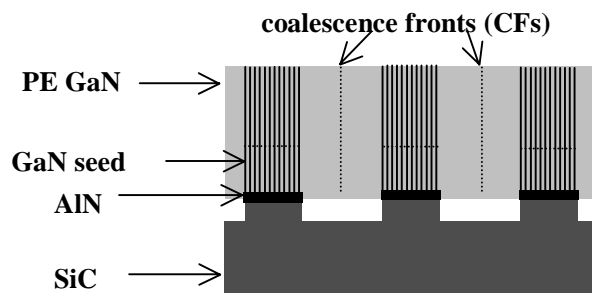


**Fig1.** Schematic diagram of the pendeo-epitaxial layer. Dislocations are shown by vertical lines.
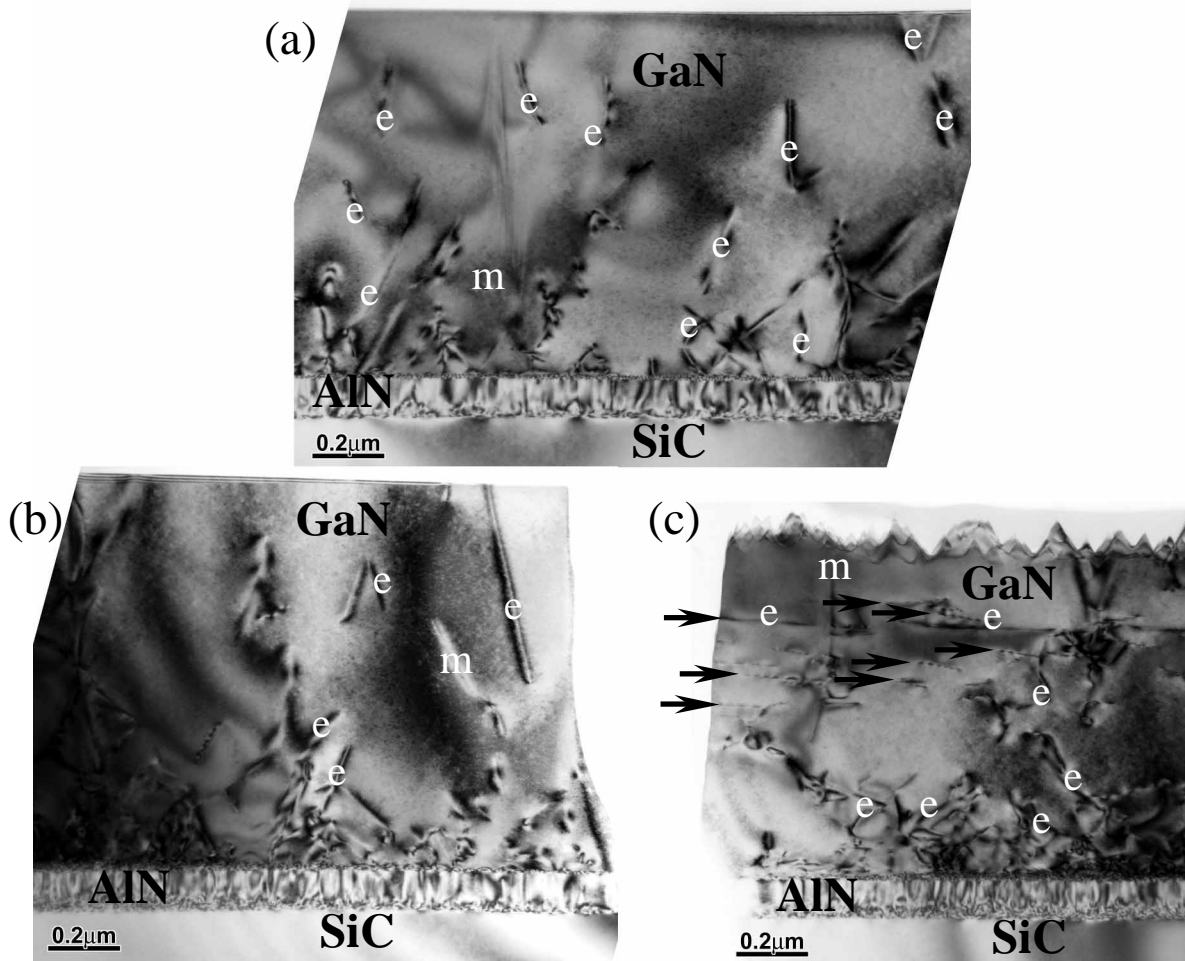
**Fig.2.** Bright field images obtained in two beam condition with $g=\underline{11}20$. (a) as deposited, (b) etched, (c) etched and annealed samples. Some edge (e) and mixed (m) dislocations are marked. Note appearance of edge dislocations with dislocation lines close to GaN c-plane in the annealed sample (marked by arrows).

56.1° formed between ($\underline{1}011$) and ($10\underline{1}1$) planes in GaN. It is concluded that walls of the pit are formed by the set of six $\{10\underline{1}1\}$ planes making an inverse pyramid with six fold symmetry. Such V-shaped inverted pyramids are often observed on the surface of GaN layers [11]. It was suggested [11] that growth on $\{10\underline{1}1\}$ planes is the slowest compared to on $\{1\underline{1}00\}$, $\{1\underline{2}10\}$ and $\{000l\}$. It appears from this study that the etching process on these planes is faster. From the measurement of the average layer thickness (0.9µm) it appears that some loss of material took place during annealing. It is noted that a degradation of the stripe surface is only observed for the annealing but not for PE growth. In the latter case the decomposition of the GaN is overcompensated by the growth such that an atomically smooth surface is maintained [9].

Dislocation density in the annealed sample remained almost the same at the area close to the AlN buffer layer ($23 \times 10^8 \mathrm{cm}^{-2}$, $0 < t < 0.5 \mu m$); however it increased to the $17 \times 10^8 \mathrm{cm}^{-2}$ for the thickness region of $0.5 < t < 0.9 \mu m$. The increase in dislocation density occurred mainly due to a new type of defects observed in the upper part of the sample (marked by arrows on Fig.2c). The dislocations are in contrast for $g=\underline{11}20$ and out of contrast for $g=0002$ which suggest that they are edge dislocations with dislocation line lying in the GaN c-plane with Burgers vectors
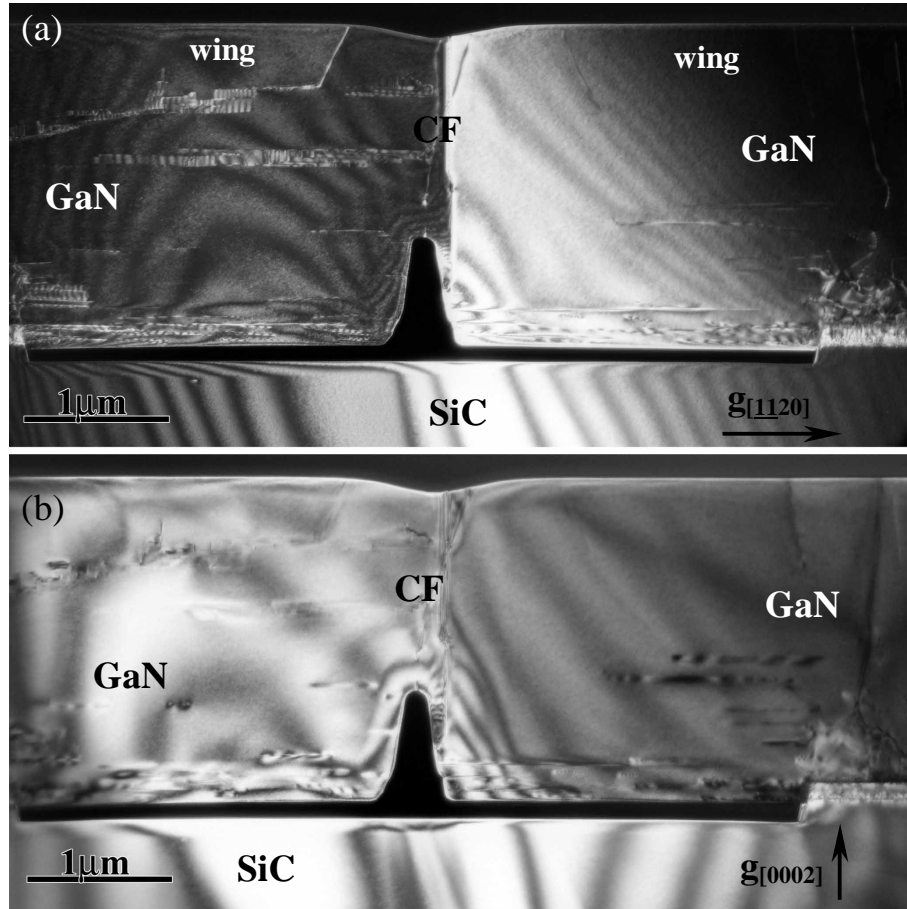
**Fig.3.** Cross-section dark field images from the PE sample taken with *g*-vectors (a) *g*=$\underline{1}1\underline{2}0$, different contrast showing an evidence of wings misorientation, (b) *g*=0002. There is a crack formation at the coalescence front (CF).

*b*=1/3<$\underline{1}1\underline{2}0$>.

The final sample, with PE growth performed, had a total GaN thickness of 2.26µm. After the growth the sample surface was smooth. There were no V-shape pits observed. This suggests that any V-shaped defects formed previously were overgrown. Dark field images (Fig.3a,b) taken with two perpendicular *g*-vectors ($\underline{1}1\underline{2}0$ and 0002) revealed a characteristic void and crack formation at the CF. However, it is not clear yet if the cracks were formed during growth or during the TEM sample preparation procedure. Almost all dislocations observed in wing areas were edge type. The dislocation density in wing regions was about $8.3 \times 10^7 \text{cm}^{-2}$, except for the highly defected area ~250nm from the bottom surface of the overgrown material. Change of contrast along defect lines from dark to bright on the image obtained with *g*=$\underline{1}1\underline{2}0$ suggests that this is a dislocation with line direction lying in the GaN c-plane at a specific angle to [$1\underline{1}00$]. Taking into account absence of contrast from the dislocations in *g*=0002 and six fold symmetry of (0001) GaN one can conclude that this is Shockley partial dislocations bordering a stacking fault in the crystal. Observation of such defects in two beams condition for *g*=$\underline{1}010$ revealed the contrast from stacking faults. Some of them propagate from the sidewall inside a wing and are terminated by the above-mentioned Shockley partial dislocations with Burgers vectors *b*=1/3<$1\underline{1}00$> [12,13]. Some probably begins at AlN/GaN interface to accommodate mismatch in *c* lattice parameters.
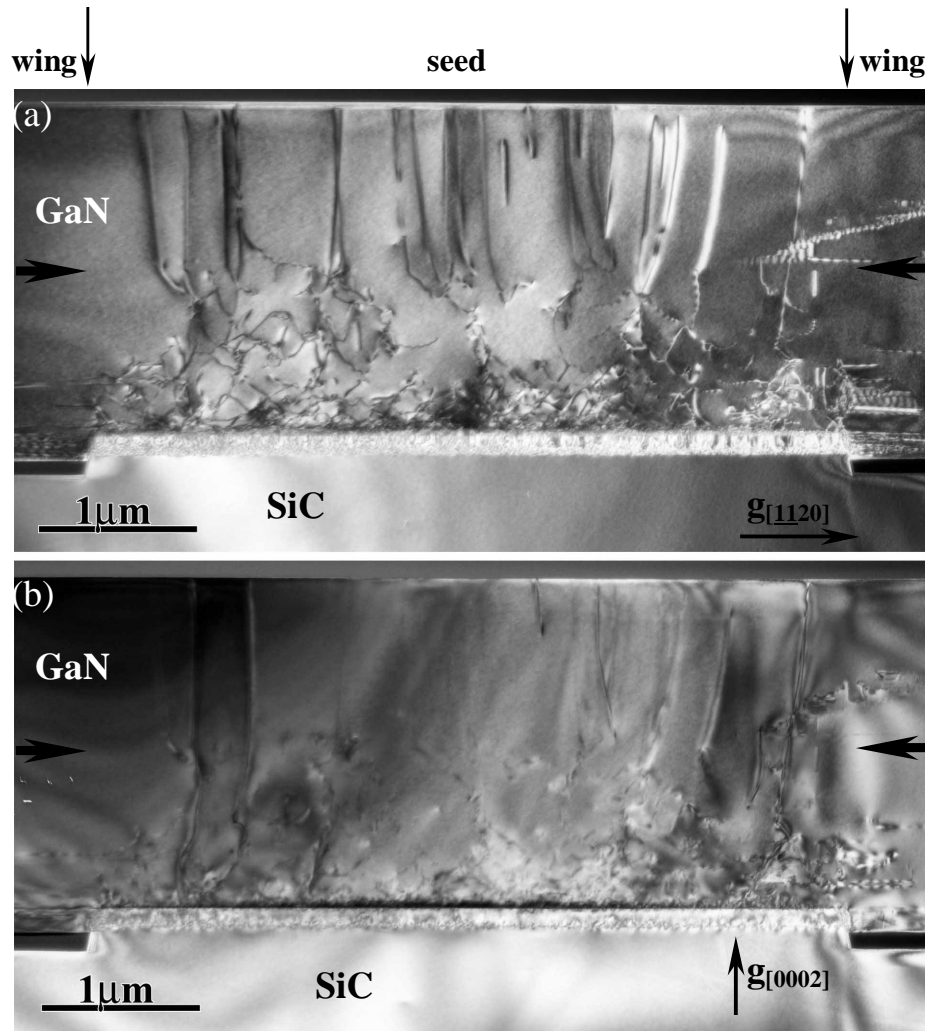
**Fig.4.** Cross-section dark field images from the PE sample (seed region) taken with two perpendicular $g$-vectors: (a) $g=1\underline{1}20$, edge and mixed dislocations are visible, (b) $g=0002$, screw and mixed dislocations are visible. Large arrows show the surface level for the seed-stripe before overgrowth.

   The difference in contrast between the wings in Fig.3a suggests an angular misorientation. A change of contrast was also observed between the seed and the wing (see right side of the Fig.3a). This suggests that the wing bends with respect to the seed. From this study it is clear that there are two misorientations in the sample: one between the two wings and the second between a wing and adjacent seed-stripe region. X-ray diffraction indicated a wing tilt of 0.26° for this sample.
   Since the surface of the final sample, where PE growth was completed, was smooth over the seed-stripe suggests that during the growth V-shaped pits were overgrown. Based on dark field images taken with two perpendicular $g$-vectors (Fig.4a,b) there are dislocations in the overgrown seed region. Most of them are edge dislocations. The density of the edge dislocations was about $8.0 \times 10^8 \text{cm}^{-2}$, which is approximately one order of magnitude more, than the density of dislocations in the wing areas ($8.3 \times 10^7 \text{cm}^{-2}$). Two large arrows show the level of 1µm thick GaN seed before PE process. One can clearly see that the edge dislocations appear approximately at

the level where V-shaped pits were observed in the annealed sample. Therefore, these pits probably act as a source of edge dislocations. Formation of additional edge dislocations, e.g. inserting (or removing) of half planes in the upper part of the layer grown over the seed would lead to the deformation (such as tilt) of the adjacent wing areas. Since this deformation might be different in different areas of the sample tilt/twist can be introduced between the wings and also between the seeds and the wings. Then it might affect tilt arising due to elastic relief of thermally induced stress described in [8].

It should be stressed that edge dislocations observed earlier in the annealed sample with etched seed-stripes (marked by arrows in Fig.2c) were not present in seed-stripe regions of PE overgrown sample.

In summary, we investigated a set of samples from at each step of the PE process. It was found that etching of a GaN layer into seed-stripes does not change the layer's surface morphology, or the dislocation distribution. However, annealing of the etched sample at a temperature comparable to GaN growth temperature resulted in formation of V-shapes pits on the surface and edge dislocations in the upper part of the GaN layer. V-shape pits probably acts as a source of edge threading dislocations observed over the seed-stripe region in the PE overgrown sample. These edge dislocations may influence observed misorientation at seed/wing and wing/wing interfaces.

## ACKNOLEDGMENTS

## REFERENCES

1. O. Nam, T. Zheleva, M. Bremser, and R.F. Davis, J. Electron Matter. **27**, 333 (1998)
2. H. Marhard, X.H. Wu, J.P. Ibbetson, P.T. Fini, P. Kozodoy, J.S. Peck, S.P. DenBaars, and U.K. Mishra, Appl. Phys. Lett. **73**, 747 (1998)
3. Z. Liliental-Weber and D. Cherns, J. Appl. Phys., **89**, 7833 (2001)
4. T. Zeleva, S. Smith, D. Thomson, K. Linthicum, P. Rajagopal, and R.F. Davis, J. Electron Matter. **28**, L5 (1999)
5. K. Linthicum, T. Gehrke, D. Thomson, E. Carlson, P. Rajagopal, T. Smith, D. Batchelor, and R.F. Davis, Appl. Phys. Lett. **75**, 196 (1999)
6. Z. Liliental-Weber, J. Jasinski, D. Cherns, M. Baines, and R. Davis, Mat. Res. Soc. Symp. Proc. **693**, 309 (2002)
7. P.Q. Miraglia, E.A. Preble, S. Einfeldt, T. Stiles, R.F. Davis, J. Schuck, R.Grober, and U. Schwarz, Phys. Stat. Sol. (a) **188**, No.2, 729-732 (2001)
8. S. Einfeldt, A.M. Roskowski, E.A. Preble, and R. Davis, Appl. Phys. Lett. **80**, 953 (2002)
9. Amy M. Roskowski, Edward A. Preble, Sven Einfeldt, Peter M. Miraglia, Robert F. Davis, IEEE J. Quantum Electron. **38**, 1006 (2002).
10. J. Jasinski and Z. Liliental-Weber, J. Electron. Mat. **31**, 429 (2002)
11. Z. Liliental-Weber, Y. Chen, S. Ruvimov, and J. Washburn, Phys Rev. Lett. **79**, 2835 1997
12. L.T. Romano, EMIS Datareviews Series No.23, INSPEC, London, UK, p.221 (1999)
13. L.T. Romano, B.S. Krusor, R.J. Molnar, Appl. Phys. Lett., **71**, 2283 (1997)